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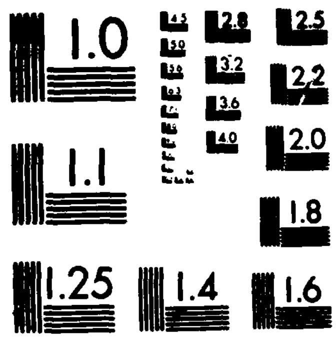

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Monthly Status Report No. 7

DEVELOPMENT OF TAS CRYSTALS FOR HARMONIC GENERATION

T. Henningsen, N. B. Singh, M. Garbuny,
K. D. Grimmett-Dawson, and K. C. Yoo
Applied Physics Department

May 11, 1987

Covering the Period
March 30, 1987 to April 26, 1987

Naval Research Laboratory
Contract No. N00014-86-C-2152

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Westinghouse R&D Center
1310 Beulah Road
Pittsburgh, Pennsylvania 15235

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DEVELOPMENT OF TAS CRYSTALS FOR HARMONIC GENERATION

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DEVELOPMENT OF TAS CRYSTALS FOR HARMONIC GENERATION

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Westinghouse R&D Center
Pittsburgh, Pennsylvania 15235

In this report we discuss some of the test results for the crystals in the second delivery on the contract. N 106, 6.2 cm long, and N 203, 6.9 cm long, are designed and AR-coated for second harmonic generation (SHG) at 19 degrees to the c-axis, and N 209, 4.9 cm long, is designed for third harmonic generation at 20 degrees to the c-axis. Table 1 includes the growth parameters for these three crystals. The crystals in this delivery are the best ones fabricated from the TAS material grown during the first nine months of the program. They were optically inspected using the infrared camera system described in the contract reports. This system evaluates the crystals at a wavelength centered at 1.4 µm close to the absorption edge of the TAS material. All three crystals show good optical image quality for o-rays, while some distortions are visible with e-rays. None of the crystals show defects, such as cracks or inclusions within the central 2 cm diameter of the crystal aperture. The birefringent interference patterns for the crystals are very complex, which we believe is indicative of a subgrain structure in the material. Overall the crystals are of better quality than the ones in the first delivery on the contract, and, with the exception of crystal N 75, also better than the ones delivered on the previous contract. Table 1 shows some of the SHG performance data for the three crystals. Also shown are the extrapolated efficiencies that we would expect from operation with a 60 nsec pulse length and a fluence of 1 joule/ cm^2. These extrapolated parameters correspond to the operating characteristics of the laser presently under construction at Westinghouse. Crystals N 106 and N 203 are coated with our standard SHG AR coating on both ends. The characteristics of the three layer BaF_2, PbF_2, ZnSe coating were described in the contract phase I report, May 4, 1984.

A reflectivity of less than 1% is being obtained at 9.6 and 4.8 µm. Each crystal is mounted in a cell sealed with AR coated ZnSe windows obtained from Two Six Corp. The transmission curve for these windows is shown in figure 1. The transmission of the assembly, which consists of the TAS crystal and two windows, was measured to be 88.2 % for N 106 and 93.8 % for N 203.

N 209 is coated for third harmonic generation. Figure 2 shows the 3.2 µm output power as a function of 9.6 µm input power to the second harmonic driver, N 203. Figure 3 shows the same output as a function of the 4.8 µm output from N203. The crystal is coated with the AR coating for 4.8 and 9.6 µm described above on the input surface, and has a mixed fluoride, 60% CeF_3 + 40% ZnSe, quarter wave coating for 3.2 µm on the output end. The reflectivity of this coating is less than 0.2 % at 3.2µm.

A small gas space is maintained between the ends of the crystal and the windows. The entrance aperture formed by the cap holding the window is a little smaller than the useful aperture of the TAS crystal, which in turn is a little smaller than the exit aperture of the window cap. This is done to help prevent the trapping of laser radiation inside the assembly which potentially might damage the crystal or release smoke from its mountings. The crystal is soft mounted with two rubber pads near the center of the crystal. This system supports the crystal with a minimum of strain and permits normal handling. The crystal is preferably used and stored in the horizontal position to avoid contact between the ends of the crystal and the windows. The cells used for these three crystals have an increased gas space of about 6 mm between the window and the end of the crystal, as compared to 1 mm for the older assemblies. This design change provides a better chance for saving the window ($600) should the surface on the TAS crystal be damaged during tests. If the testing is stopped as soon as damage is detected, the crystal can be repolished and recoated, and the window cleaned.

The crystals were tested with a Lumonix CO_2 laser model 203. The wavelength was tuned to the P(24) transition of the (00^01-02^00) CO_2 band at 9.6 µm. An intracavity aperture of 16 mm diameter was used to obtain a TEM_{00} mode. The beam was polarized with its E-vector in the vertical plane and operated in the longitudinal multimode.

The test arrangement and orientations of the laser beam and crystal are shown in figure 4. In the presently used TAS crystal growth direction, SHG phase match is achieved when the laser beam enters as ordinary ray in a b-c plane at 19^0 with respect to the c-axis of the crystal. In figure 4, the plane of the drawing contains the b- and c-axis as well as the direction of the laser beam. The beam polarization is normal to the plane of the drawing. The line on the crystal holder should be on top for the crystal to function as indicated in figure 4. In the crystal growth procedure, the boule axis can normally be held to the desired orientation within 1^0 -2^0. Thus fine tuning of the phase match angle is necessary. This is accomplished by turning the crystal from normal beam incidence around an axis parallel to the beam polarization.

The spending during the month of April was $72,591 for a total of $857,830 since the start of the program June 18, 1986. This represents 101% of the incremental funds of $844,000 allocated to the program for the period June 18,1986 to September 30, 1987. Total funding of the program to the completion date June 18, 1988 is $ 1,312,398. The program is currently on schedule, and in most respects meeting its objectives.

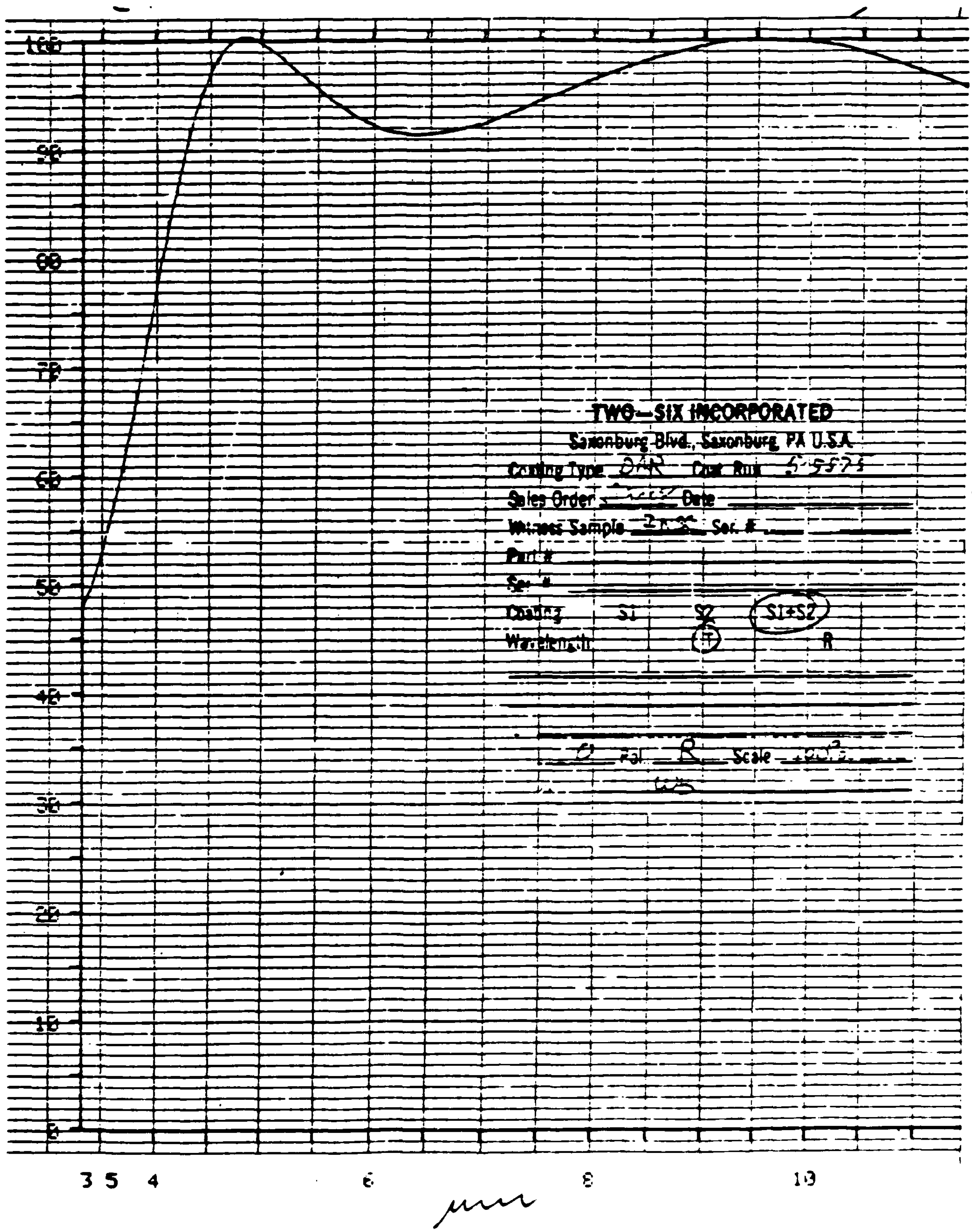

Figure 1. AR coating for 4.8 and 9.6 µm on ZnSe windows.

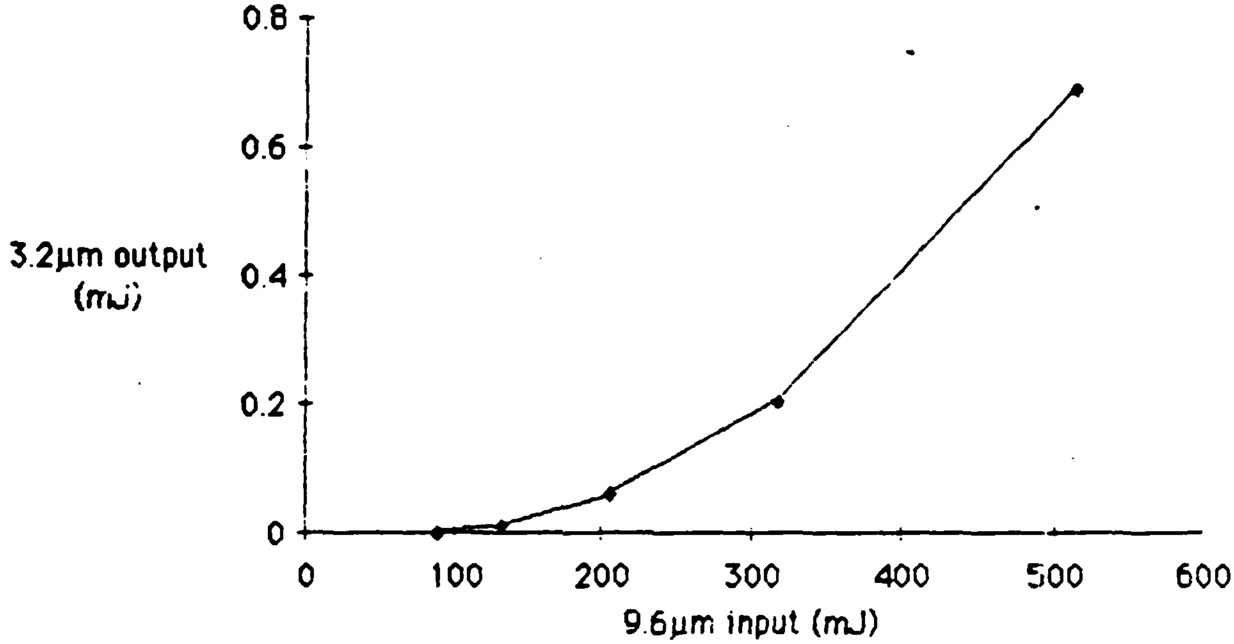

Figure 2. Third harmonic output energy, 3.2 µm, as a function of fundamental input energy, 9.6µm, to the second harmonic generator.

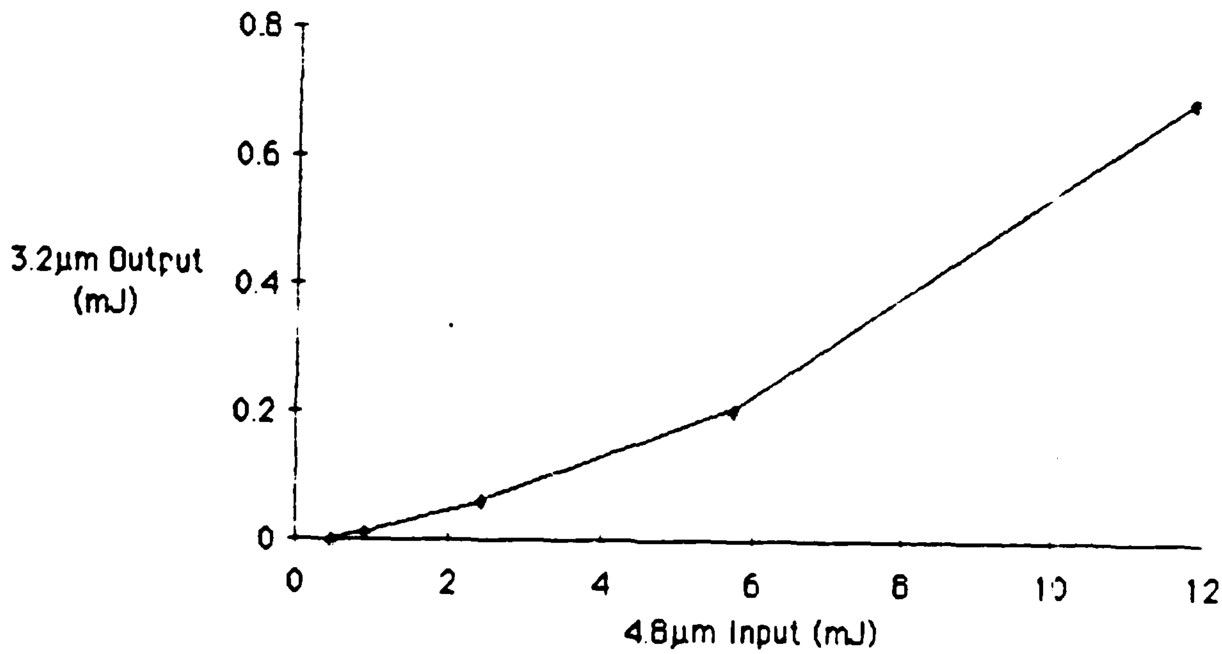

Figure 3. Third harmonic output energy, 3.2 µm, as a function of second harmonic input energy, 4.8 µm, from the second harmonic generator.

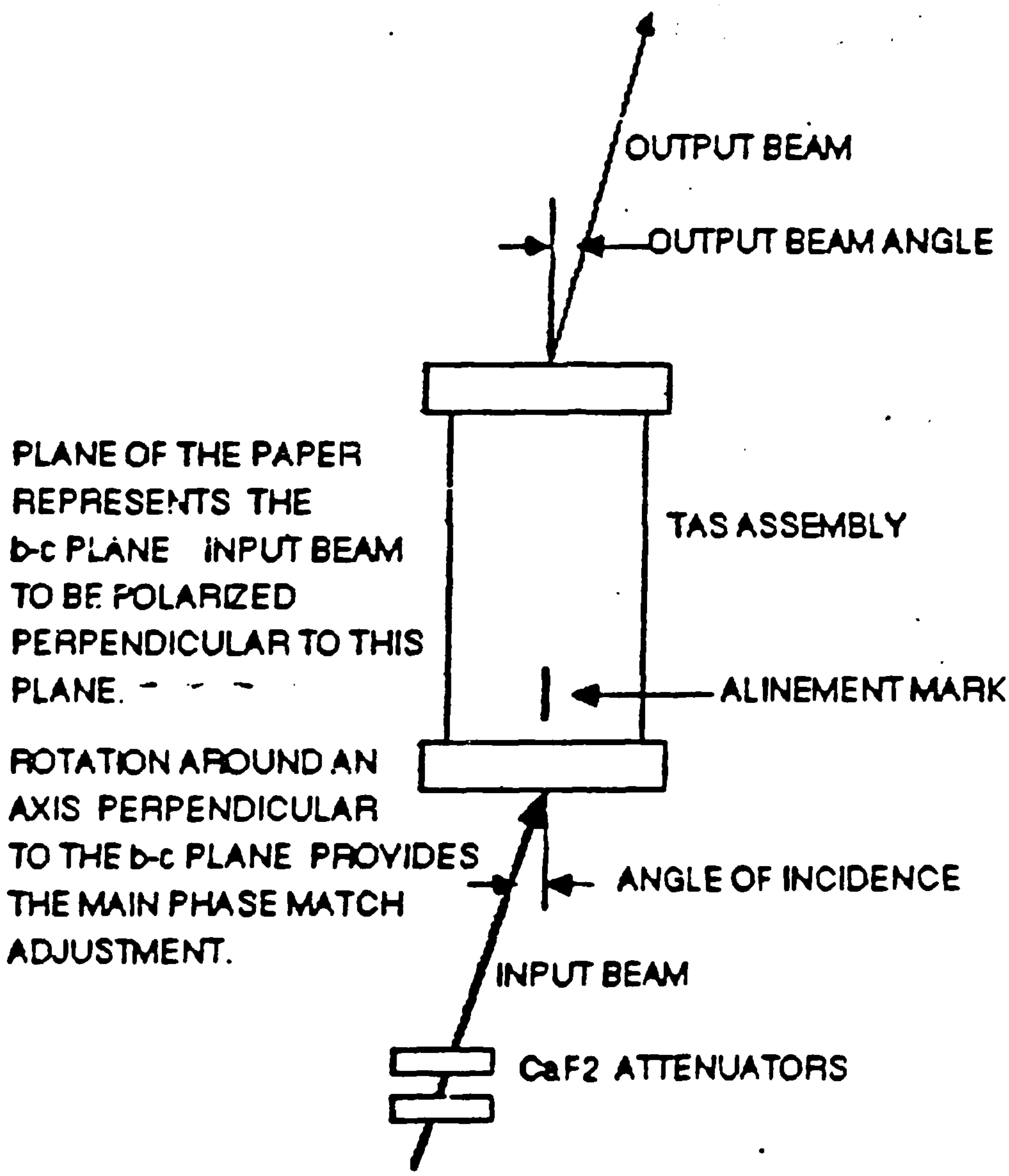

Figure 4. The test arrangement and orientations of the laser beam and crystal .

Table 1. Summary of measured SHG parameters.

Measured Values:										
Crystal I. D.	Length cm	Apt. Eff.	Power J/cm2	Extrap. 60ns pulse Efficiency @1 J/cm2	SHG Beam Divergence	Full Beam Efficiency	Power Joules	Efficiency @1.0 Joule	AR Coated	Comments
N203II(23-98)coated	6.9	5.14	0.402	43.7	3.7mrad	2.57	0.454	5.4	YES	AR-coated for shipment to the NAVY (SHG)
N106(36-104) coated	6.2	2.68	0.524	22.0	3.9mrad	1.5	0.504	3	YES	AR-coated for shipment to the NAVY (SHG).
N209(84-140) coated	4.9	2.16	0.401	22.8	3.4mrad	1.5	0.393	3.9	YES	AR-coated for shipment to the NAVY (THG).

Monthly Status Report No. 8

DEVELOPMENT OF TAS CRYSTALS FOR HARMONIC GENERATION

T. Henningsen, N. B. Singh, M. Garbuny,
K. D. Grimmett-Dawson, and K. C. Yoo
Applied Physics Department

June 12, 1987

Covering the Period
April 27, 1987 to May 31, 1987

Naval Research Laboratory
Contract No. N00014-86-C-2152

Westinghouse R&D Center
1310 Beulah Road
Pittsburgh, Pennsylvania 15235

DEVELOPMENT OF TAS CRYSTALS FOR HARMONIC GENERATION

T. Henningsen, N. B. Singh, M. Garbuny, K. D. Grimmett-Dawson, and K. C. Yoo.
Westinghouse R&D Center
Pittsburgh, Pennsylvania 15235

The growth of boule S-103 has been completed. Initial inspections shows the crystal to have an improved grain structure when compared with older crystals, but it still does have a subgrain structure. Further fabrication and testing will be performed during the next period. Boule S-104 was being grown using the same seeding technique that produced S-102. At the end of the growth but before the boule had been cooled to room temperature, a wire failure caused a heavy counter weight to fall and hit the furnace and exposed the boule to a very strong vibration which we later found had produced internal cracks in the boule along the growth axis. Figure 1 shows an X-ray topography picture of the boule. We see that the cross section of the boule represents a single subgrain. This is the first time this has been achieved and shows the promise of, and the need for, improved seeding techniques. The intensity variations seen in the picture are caused by surface layer effects and residual strain fields in the boule. The picture also shows the cracks caused by the furnace malfunction. The top and bottom sections of the boule were removed and discarded . The remaining section of the boule plus some extra material was used to regrow the crystal. This growth should be completed during the next reporting period.

Table 1 shows a summary of the growth and test parameters on the boules grown on this program. About 30 boules have been grown and evaluated. Each growth run was designed to examine the various parameters of the materials purification and the growth process. Table 1 lists the purification steps, growth rate, temperature gradients in the furnace, and the doping of the boules. The experiments have produced a good base of technology in these areas. Work in the next few months will be concentrated on

reducing the subgrain boundaries in the boules.

The spending during the month of May was $51,624 for a total of $909,454 since the start of the program June 18, 1986. This represents 108 % of the incremental funds of $844,000 allocated to the program for the period June 18, 1986 to September 30, 1987. Total funding of the program to the completion date June 18, 1988 is $ 1,312,398. The work on the program is being curtailed to avoid further overruns of the funds currently allocated for the program. We are presently concentrating on the testing and fabrication of the crystals scheduled for delivery June 18, 1987.

Figure 1. Single subgrain cross section of boule S-104

TABLE 1. Growth parameters for TAS

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X-tal No.	Feedstack Purification Tl	As	Se	Pregrowth Zone Refining	Pregrowth Directional Freezing	Seeded Growth Rate (cm/day)	Temperature Gradient (°C/cm)	Doping	Furnace Type ✦	Expt. Purpose
N-101	X									Precontract
N-101	X		X	N	Y	1.02	50 ~ 60	N	SF(1)	Baseline Data
N-102	X		X	N		0.9	50 ~ 60	Ag	SF(1)	Study Doping Effect
N-103	X		X	N	Y	1.24	50 ~ 60	N	SF(1)	Baseline Data
N-104	X		X	N	Y	0.91	30 ~ 35	N	MF	Test Reduced Gradient
N-105	X	X	X	N	Y	1.03	50 ~ 60	N	SF(1)	Test Purified As
N-105 (II)	X	X	X	N		0.87	45	N	MF	Regrow to Improve Quality
N-106	X	X	X	N	Y	0.9	30 ~ 35	Ag	MF	Test Purified As
N-107	X	X	X	N	Y	1.04	30 ~ 35	N	MF	Test Purified As
N-201	Regrowth Using Previous X-tal			Y (10 Passes)		1.06	50 ~ 60	N	SF(1)	To Check Effect of Zone Refining
N-202	Regrowth Using Previous X-tal			Y (10 Passes)	Y	1	30 ~ 35	N	MF	To Check Effect of Zone Refining
N-203	Regrowth Using Previous X-tal			Y (17 Passes)	N	0.73	25	Ag	SF(2)	To Check Effect of Zone Refining
N-203 (II)				N	N	0.74	25	Ag	SF(2)	Regrowth
N-204	X		X	Y (12 Passes)	N	0.98	30 ~ 35	Cu	MF	Study Doping Effect
N-205	X		X	Y (14 Passes)	N	1.02	30 ~ 35	Ag	MF	Study Doping Effect
N-206	X		X	N	Y	1	50 ~ 60	N	SF(1)	Test Effects of Tl-Purification
N-207	Under Growing									
N-208	X	X	X	Y (8 Passes)	N	0.86	30 ~ 35	Ag	SF(1)	Zone Refined Material w/Ag Dope
N-209	X		X	N	N	1	50 ~ 60	N	SF(2)	Reproduct of N-206
N-210	X	X	X	Y (8 Passes)	N	0.96	38	Cu	MF	Doping Effect
N-211	X		X	N	Y	1	38	Ag	MF	Doping Effect
N-212	X	X	X	Y (9 Passes)	N	0.72 3.7	24	N	SF(2)	Effect of 2-Zone Furnace
S-101	X		X	N	Y	1	45	N	MF	Seeding Exp.
S-102	X		X	N	Y	0.8	38	N	MF	Seeding Exp.
S-103	X		X	N	Y	0.8	38	N	MF	Seeding Exp.
S-104	X		X	N	Y	0.8	32	N	MF(2)	Seeding Exp.
N-213	x		x	Y(15 P.)	N	Waiting for Regrowth				
N-214	x		x	Y(7 P.)	N	1.1	20	N	SF(2)	For 4th Harmonic Generation w/ 4x4 mm Seed
S-104(II)	Regrowth using S-104					0.8	32	N	MF	Regrowth
N-107(II)	Regrowth using N-107					1.1	20	N	SF(2)	Necking Effect

Legend:

- "X" Indicates that Pregrowth Purification of the As-Received Element was Performed
  "Y" Indicates Yes
  "N" Indicates No
- ✦ MF - Furnace Reverses, Crystal Stationary. SF (# Zones) - Stationary Furnace Crystal Reverses (1 or 2 Hot Zones)

Monthly Status Report No. 9

DEVELOPMENT OF TAS CRYSTALS FOR HARMONIC GENERATION

T. Henningsen, N. B. Singh, M. Garbuny,
K. D. Grimmett-Dawson, and K. C. Yoo
Applied Physics Department

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Naval Research Laboratory
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Westinghouse R&D Center
1310 Beulah Road
Pittsburgh, Pennsylvania 15235

DEVELOPMENT OF TAS CRYSTALS FOR HARMONIC GENERATION

T. Henningsen, N. B. Singh, M. Garbuny, K. D. Grimmett-Dawson, and K. C. Yoo.
Westinghouse R&D Center
Pittsburgh, Pennsylvania 15235

In the previous monthly report, no 8, we discussed the growth of boule S-104 which, despite a furnace malfunction that caused cracks in the crystal, for the first time demonstrated the growth of TAS material free of subgrain boundaries. The regrowth of this boule was completed during this period. This time we experienced no problems with the furnaces. However, x-ray topography shows that the resulting boule had multiple subgrain boundaries which could be traced back through the necked down area in the seed holder and into the seed. Birefringent interferometry shows fringe patterns in the different subgrain regions, but with orientations that vary from one subgrain to the next. We conclude that the boule grain structure is better than the structures seen in most of the TAS crystals grown on the program but that the seeding arangement in this case did propagate multiple subgrains. The overall experience from the S series crystals, which all use the neck down technique in the seed holder, is that this procedure does significantly reduce the number of subgrains but does not by itself insure that only a single subgrain will propagate.

In order to further reduce the probability of the subgrain boundary propagation through the neck, it is necessary to prepare the seeds without any subgrains, or if the subgrains can not be avoided, to prepare seeds with the subgrain boundaries oriented perpendicular to the direction of boule growth. Through many x-ray topography observations of the micro-defect structures of the TAS crystals, we have found that the subgrain boundaries form along the boule growth direction. One of the examples is shown in the Berg-Barrett x-ray topograph of Figure 1. Therefore, we should be able to

obtain seeds with the desired subgrain orientations from boules grown in a direction perpendicular to the directions used for the harmonic generators used on this program. The seed is then prepared from these special seed boules with its long dimension (the C-19 direction for the SHG crystals) perpendicular to the seed boules growth orientation. This is one of the approaches to the seeding problem currently being pursued on the program.

During a meeting with the contract monitor at NRL, June 18,1987, it was agreed that the most important problem at the present time was to improve the crystal seeding in preference over the scheduled delivery of three crystals in June 1987. It was decided to delay two of the crystals to later in the program when we expect better material to be available. The third crystal to be delivered now is a fourth harmonic generator grown at 28 degrees to the C-axis. Tentatively, we identified N-214 as suitable for this purpose. The crystal had performed well in the initial screening test. However, during the final surface preparation, the crystal developed surface cracks originating at strain points on the cylindrical surface. While the crystal still has a clear aperture of about 17 mm, these cracks may eventually propagate across the aperture of the crystal. We have, therefore, proceeded to fabricate another 28 degree crystal, PR-2, for fourth harmonic generation. This crystal is currently being coated.

The spending during the month of June was $ 9646 for a total of $919,100 since the start of the program June 18, 1986. This represents 109 % of the incremental funds of $844,000 allocated to the program for the period June 18, 1986 to September 30, 1987. Total funding of the program to the completion date June 18, 1988 is $1,312,398.

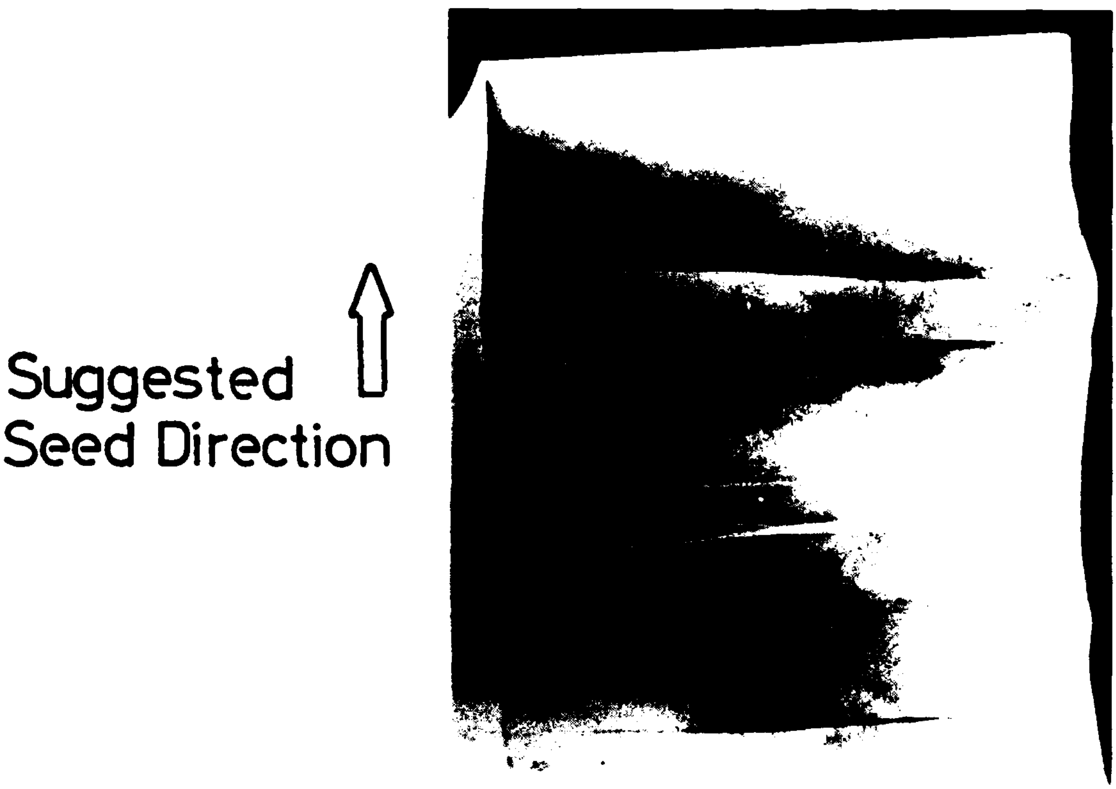

Figure 1. An x-ray topograph taken from a TAS single crystal shows a number of subgrain boundaries oriented along the crystal growth direction.

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